

Two-magnon Raman scattering in spin-ladders with exact singlet-rung ground state

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Abstract

Using coordinate Bethe ansatze we construct two-magnon states for the suggested by A. K. Kolezhuk and H.-J. Mikeska family of spin-ladder models with exact singlet-rung vacuum. The explicit formula for zero-temperature Raman scattering cross section is derived. The corresponding line-shapes are strongly asymmetric and their singularities originate only from bound states. This form of a line-shape is in a good correspondence to the experimental data.

1 Introduction

Raman scattering in spin-ladders was studied in a number of papers (see [1]-[9] and references therein). The obtained experimental data was analyzed by several theoretical approaches [3],[7],[8]. However in none of these papers the exact formula for the Raman cross section was used. In the present paper we obtain the *exact* formula for the special class of spin-ladder models with exact singlet-rung vacuum. This family of models was first suggested in [10]. The corresponding Hamiltonian \mathcal{H} has the following form:

$$\mathcal{H} = \sum_{n=-\infty}^{\infty} H_{n,n+1}, \quad (1)$$

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where

$$H_{n,n+1} = H_{n,n+1}^{stand} + H_{n,n+1}^{frust} + H_{n,n+1}^{cyc} + H_{n,n+1}^{norm}, \quad (2)$$

and

$$\begin{aligned} H_{n,n+1}^{stand} &= \frac{J_{\perp}}{2}(\mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n} + \mathbf{S}_{1,n+1} \cdot \mathbf{S}_{2,n+1}) + J_{\parallel}(\mathbf{S}_{1,n} \cdot \mathbf{S}_{1,n+1} + \mathbf{S}_{2,n} \cdot \mathbf{S}_{2,n+1}), \\ H_{n,n+1}^{frust} &= J_{frust}(\mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n+1} + \mathbf{S}_{2,n} \cdot \mathbf{S}_{1,n+1}), \\ H_{n,n+1}^{cyc} &= J_c((\mathbf{S}_{1,n} \cdot \mathbf{S}_{1,n+1})(\mathbf{S}_{2,n} \cdot \mathbf{S}_{2,n+1}) + (\mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n})(\mathbf{S}_{1,n+1} \cdot \mathbf{S}_{2,n+1}) \\ &\quad - (\mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n+1})(\mathbf{S}_{2,n} \cdot \mathbf{S}_{1,n+1})), \\ H_{n,n+1}^{norm} &= J_{norm}I. \end{aligned} \quad (3)$$

Here $\mathbf{S}_{i,n}$ ($i = 1, 2$; $n = -\infty \dots \infty$) are spin- $\frac{1}{2}$ operators associated with cites of the ladder and I is an identity matrix. The auxiliary term $H_{n,n+1}^{norm}$ in (2) is need only for normalization to zero the lowest eigenvalue of the 16×16 matrix H of rung-rung interaction.

It was shown in [10] that when the following conditions

$$\begin{aligned} J_{frust} &= J_{\parallel} - \frac{1}{2}J_c, \quad J_{norm} = \frac{3}{4}J_{\perp} - \frac{9}{16}J_c, \\ J_{\perp} &> 2J_{\parallel}, \quad J_{\perp} > \frac{5}{2}J_c, \quad J_{\perp} + J_{\parallel} > \frac{3}{4}J_c. \end{aligned} \quad (4)$$

are satisfied then the lowest (zero eigenvalue) eigenstate of H is $w \otimes w$, where w is the rung-singlet state. In this case the ground state of the Hamiltonian (1) has the simple tensor-product form:

$$|0\rangle = \prod_n \otimes w_n. \quad (5)$$

In order to obtain the full spectrum of H we shall also define the following triplet states:

$$f_n^k = (\mathbf{S}_{1,n}^k - \mathbf{S}_{2,n}^k)w_n, \quad (\mathbf{S}_{1,n}^j + \mathbf{S}_{2,n}^j)f_n^k = i\varepsilon_{jkm}f_n^m. \quad (6)$$

All other eigenstates of H are separated into the following sectors: singlet $f^k \otimes f^k$, triplet $\varepsilon_{ijk}f^j \otimes f^k$, quintet $t_{ijkl}f^j \otimes f^k$ with eigenvalues: $\varepsilon_0 = J_{\perp} - 2J_{\parallel}$, $\varepsilon_1 = J_{\perp} - J_{\parallel} - \frac{1}{4}J_c$, $\varepsilon_2 = J_{\perp} + J_{\parallel} - \frac{3}{4}J_c$, and two triplets $w \otimes f^k \pm f^k \otimes w$ with eigenvalues: $\varepsilon_{\pm} = \frac{1}{2}(J_{\perp} - \frac{3}{2}J_c \pm J_c)$. Here $t_{ijkl} = \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}$.

The Hamiltonian (1)-(3) commutes with the following magnon number operator $\mathcal{Q} = \sum_n Q_n$, where $Q_n = \frac{3}{4}I + \mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n}$ is the associated with the n -th rung projection operator on triplet states.

2 The two-magnon states

Corresponding to (1)-(4) one-magnon states were obtained in [10]. Suggesting the following Bethe form for two-magnon states $|S, \beta\rangle$ (where S is the total spin and β the list of additional parameters):

$$|0; \beta\rangle = \sum_{m=-\infty}^{\infty} \sum_{n=m+1}^{\infty} a_0(m, n; \beta) \dots w_{m-1} f_m^j w_{m+1} \dots w_{n-1} f_n^j w_{n+1} \dots, \quad (7)$$

$$|1; \beta\rangle_i = \sum_{m=-\infty}^{\infty} \sum_{n=m+1}^{\infty} a_1(m, n; \beta) \varepsilon_{ijk} \dots w_{m-1} f_m^j w_{m+1} \dots w_{n-1} f_n^k w_{n+1} \dots, \quad (8)$$

$$|2; \beta\rangle_{ij} = \sum_{m=-\infty}^{\infty} \sum_{n=m+1}^{\infty} a_2(m, n; \beta) t_{ijkl} \dots w_{m-1} f_m^k w_{m+1} \dots w_{n-1} f_n^l w_{n+1} \dots \quad (9)$$

we obtain in standard way [11] the following Schrödinger equation:

$$\begin{aligned} \frac{J_c}{2} [a_S(m-1, n; \beta) + a_S(m+1, n; \beta) + a_S(m, n-1; \beta) + a_S(m, n+1; \beta)] \\ + (2J_{\perp} - 3J_c) a_S(m, n; \beta) = E a_S(m, n; \beta), \end{aligned} \quad (10)$$

and Bethe condition for amplitudes:

$$2\Delta_S a_S(n, n+1; \beta) = a_S(n, n; \beta) + a_S(n+1, n+1; \beta). \quad (11)$$

Here $\Delta_S = \frac{\varepsilon_S - \varepsilon_+ - \varepsilon_-}{\varepsilon_+ - \varepsilon_-}$.

For each S the Eq. (11) has two solutions. The scattering solution:

$$a_S^{scatt}(m, n; k_1, k_2) = C_{S,12} e^{i(k_1 m + k_2 n)} - C_{S,21} e^{i(k_2 m + k_1 n)}, \quad (12)$$

with $C_{S,ab} = \cos \frac{k_a + k_b}{2} - \Delta_S e^{i \frac{k_a - k_b}{2}}$, and the bound solution:

$$a_S^{bound}(m, n; u) = e^{i u (m+n) + v(m-n)}, \quad (13)$$

where the real parameters $v \geq 0$ and $-\pi < u \leq \pi$ satisfy the following condition:

$$\cos u = \Delta_S e^{-v}. \quad (14)$$

From (14) and nonnegativity of v follows that

$$|\cos u| \leq |\Delta_S| \leq e^v. \quad (15)$$

The corresponding to (12) and (13) eigenvalues are:

$$E_S^{scatt}(k_1, k_2) = 2J_{\perp} - 3J_c + J_c (\cos k_1 + \cos k_2), \quad (16)$$

$$E_S^{bound}(u) = 2J_{\perp} + (\Delta_S - 3) J_c + \frac{J_c}{\Delta_S} \cos^2 u. \quad (17)$$

As we see from (12) and (13) the translation invariant states correspond to $a_S^{scatt}(m, n; k, -k)$, $a_S^{bound}(m, n; 0)$ and $a_S^{bound}(m, n; \pi)$.

3 Calculation of Raman cross section

Following Sugai [2] we shall consider only the case when the incident and scattered light have parallel polarization directions both lying in the plane of the ladder and forming an angle θ with respect to vertical bonds (rungs). The zero-temperature two-magnon Raman scattering cross section as a function of frequency and θ can be expressed using Fermi's golden rule [3],[4]:

$$I(\omega, \theta) = \lim_{N \rightarrow \infty} \frac{2\pi}{2N + 1} \sum_{\alpha} |\langle \alpha | \mathcal{H}^R(\theta) | 0 \rangle|^2 \delta(\omega - E_{\alpha}), \quad (18)$$

where $2N + 1$ is the number of rungs. Within the Fleury-Loudon-Elliott approach the effective Raman Hamiltonian $\mathcal{H}^R(\theta)$ have the following form [1],[5] (we also take into account interactions across diagonals):

$$\mathcal{H}^R(\theta) = A_{leg} \cos^2 \theta \mathcal{H}^{leg} + A_{diag} (\cos^2(\theta + \gamma) \mathcal{H}^{d_1} + \cos^2(\theta - \gamma) \mathcal{H}^{d_2}) + A_{rung} \sin^2 \theta \mathcal{H}^{rung}. \quad (19)$$

Here A_{leg} , A_{diag} and A_{rung} are constants and γ is the angle between rung and diagonal directions. Operators \mathcal{H}^{rung} , \mathcal{H}^{leg} and \mathcal{H}^{d_1} , \mathcal{H}^{d_2} are the following:

$$\mathcal{H}^{rung} = \sum_n \mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n}, \quad \mathcal{H}^{leg} = \sum_{i,n} \mathbf{S}_{i,n} \cdot \mathbf{S}_{i,n+1}, \quad \mathcal{H}^{d_{1(2)}} = \sum_n \mathbf{S}_{1(2),n} \cdot \mathbf{S}_{2(1),n+1}. \quad (20)$$

Expressing \mathcal{H}^{leg} , \mathcal{H}^{d_1} , \mathcal{H}^{d_2} and \mathcal{H}^{rung} from the auxiliary operators:

$$\mathcal{H}^{\pm\pm} = \sum_n (\mathbf{S}_{1,n} \pm \mathbf{S}_{2,n}) \cdot (\mathbf{S}_{1,n+1} \pm \mathbf{S}_{2,n+1}). \quad (21)$$

and taking into account the Eq. (6) we represent $I(\omega, \theta)$ in the factorized form:

$$I(\omega, \theta) = \frac{1}{4} [(A_{leg} + A_{diag} \sin^2 \gamma) \sin^2 \theta + A_{diag} \cos^2 \gamma \cos^2 \theta]^2 I_0(\omega), \quad (22)$$

where

$$I_0(\omega) = \lim_{N \rightarrow \infty} \frac{2\pi}{2N + 1} \sum_{\alpha} |\langle \alpha | \mathcal{H}^{--} | 0 \rangle|^2 \delta(\omega - E_{\alpha}). \quad (23)$$

Formula (22) expresses the polarization angle dependence of Raman cross section however it may be applied in a straightforward way only for $\theta = \frac{m\pi}{2}$ [6].

From the Eq. (6), translational and SU(2) invariance of \mathcal{H}^{--} follows that only translation invariant singlet two-magnon states contribute to the formula (23). Separating the

contributions from scattering and bound states we obtain:

$$I_0^{scatt}(\omega) = \lim_{N \rightarrow \infty} \sum_k \frac{|\sum_{n=-N}^N a(n, n+1; k, -k)|^2}{\sum_{n=-N+1}^N \sum_{m=-N}^{n-1} |a(m, n; k, -k)|^2} \delta(\omega - E_0^{scatt}(k, -k)), \quad (24)$$

$$I_0^{bound}(\omega) = \lim_{N \rightarrow \infty} \sum_{u=0,\pi} \frac{|\sum_{n=-N}^N a(n, n+1; u)|^2}{\sum_{n=-N+1}^N \sum_{m=-N}^{n-1} |a(m, n; u)|^2} \delta(\omega - E_0^{bound}(u)). \quad (25)$$

From (12) and (13) follows:

$$\sum_{n=-N+1}^N \sum_{m=-N}^{n-1} |a_0^{scatt}(m, n; k, -k)|^2 = 4N^2(1 - 2\Delta_0 \cos k + \Delta_0^2) + O(N), \quad (26)$$

$$|\sum_{n=-N}^N a^{scatt}(n, n+1; k, -k)| = 4N \sin k + O(1), \quad (27)$$

$$\sum_{n=-N+1}^N \sum_{m=-N}^{n-1} |a_0^{bound}(m, n; u)|^2 = \frac{2N}{e^{2u} - 1} + o(N), \quad u = 0, \pi \quad (28)$$

$$|\sum_{n=-N}^N a^{bound}(n, n+1; u)| = (2N+1)e^{-v}, \quad u = 0, \pi. \quad (29)$$

Using the substitution $\sum_k \rightarrow \frac{2N+1}{2\pi} \int_0^{2\pi} dk$ we obtain from (26)-(29) the final expressions for the cross sections:

$$I_0^{scatt}(\omega) = \frac{4\Theta(1-x^2)\sqrt{1-x^2}}{J_c(1+\Delta_0^2-2x\Delta_0)}, \quad (30)$$

$$I_0^{bound}(\omega) = \frac{2\pi}{J_c}(1-\frac{1}{\Delta_0^2})\Theta(\Delta_0^2-1)\delta(2x-\Delta_0-\frac{1}{\Delta_0}). \quad (31)$$

Here Θ is the step function and $x = \frac{\omega-2J_\perp+3J_c}{2J_c}$ is the rescaling parameter.

From (15) and (31) follows that the contribution from bound states $I_0^{bound}(\omega)$ exist only for $|\Delta_0| > 1$. The behavior of I_0^{scatt} as a function of x also essentially depends on the parameter $\Delta_0 = \frac{3}{2} - 2\frac{J_\perp}{J_c}$. When $\Delta_0 = \pm 1$ the formula (30) reduces and the line-shape has a singularity at $x = \Delta_0$. For $\Delta_0 = 1$ it lies in the top of the two-magnon continuum however for $\Delta_0 = -1$ in the bottom. For $\Delta_0 \neq \pm 1$ the cross section I_0^{scatt} is a regular function of x and has the maximum in the point $x_{max} = \frac{2\Delta_0}{\Delta_0^2+1}$.

In order to study the line-shape in more detail we shall find its inflection points. Calculating the second derivative of I_0^{scatt} with respect to x we obtain the following condition:

$$p(x, \Delta_0) = 4\Delta_0(1+\Delta_0^2)x^3 - 12\Delta_0^2x^2 - \Delta_0^4 + 6\Delta_0^2 - 1 = 0. \quad (32)$$

Since $p(\pm 1, \Delta_0) = -(1 \mp \Delta_0)^4$, the polynomial $p(x, \Delta_0)$ for $\Delta_0 \neq \pm 1$ has only 0 or 2 zeros in the interval $(-1, 1)$. From standard calculation follows that $p(x, \Delta_0)$ has the maximum $p_{max} = -\Delta_0^4 + 6\Delta_0^2 - 1$ in the point $x = 0$. It is evident now that for $p_{max} > 0$ the line-shape of I_0^{scatt} has two inflection points. From the straightforward calculation follows that $p_{max} > 0$ only for

$$\Delta_- < |\Delta_0| < \Delta_+, \quad (33)$$

where $\Delta_{\pm} = \sqrt{3 \pm 2\sqrt{2}}$ ($\Delta_- \approx 0.4142$, $\Delta_+ \approx 2.4142$). It may be easily proved in a straightforward way that $(\Delta_+ - \Delta_-)^2 = 4$, so $\Delta_+ - \Delta_- = 2$.

In the case (33) the line-shape near the x_{max} is similar to van-Hove singularity. For $\Delta_- < \Delta_0 < \Delta_+$ this "singularity" lies near the top of the two-magnon continuum however for $-\Delta_+ < \Delta_0 < -\Delta_-$ near the bottom. In both the cases the line-shape of Raman scattering is strongly *asymmetric*. The case $p_{max} < 0$ with no inflection points may be interpreted as a broad maximum. Some line shapes corresponding to different values of Δ_0 are presented in the Fig. 1.

As it follows from (16) and (17) for $\Delta_0 \rightarrow \pm 1 + 0^{\pm}$ the top (bottom) of the two-magnon continuum and the bound two-magnon state have the same energy: $2J\perp - 3J_c \pm 2J_c$. It was proposed in [3] that in this case the resonance between bound and scattering states leads to a redistribution of Raman intensity and merging of singularity. However as we see from (30) and (31) in our model this conjecture fails. Moreover the singularity in I_0^{scatt} appears only in the resonance $\Delta_0 = \pm 1$ cases.

4 Comparison with experiment and discussion

Raman scattering in MgV_2O_5 and CaV_2O_5 were reported in [9]. It was pointed that for both materials the corresponding line-shapes are strongly asymmetric and have one maximum instead of two. This fact was considered as strange and there were suggested some conjectures to interpret it. For example it was supposed in [9] that in MgV_2O_5 there is no spin-gap and the magnetic ordering is 2D, or the spin-gap is so small (about 10 cm^{-1}) that can not be observed by the used experimental resolution. The asymmetry of the line-shape for CaV_2O_5 was interpreted in [9] as originating from next-nearest neighbor interactions. In [7] it was conjectured that the second peak in the line-shape of CaV_2O_5 is not observed because it is dominated by phonon peak. In [3] it was conjectured that the asymmetry originates from resonance with two-triplet bound state.

In our paper we have demonstrated that the line-shape asymmetry in spin-ladder Raman scattering is not something strange and outstanding but may appear in a sufficiently big class of models. Of course we do not pretend that for some values of exchange parameters our toy model necessarily describes the real materials such as CaV_2O_5 or MgV_2O_5 . Nevertheless perhaps the true ground state is in some sense "close" to our idealized one (5) and we may believe that our model correctly represents some general qualitative features of real materials. In this context we emphasize that the *exactly calculated* Raman scattering line-shape may be strongly asymmetric without any additional assumptions such as next-nearest neighbor interactions, resonance with bound state or dominating by phonon peak.

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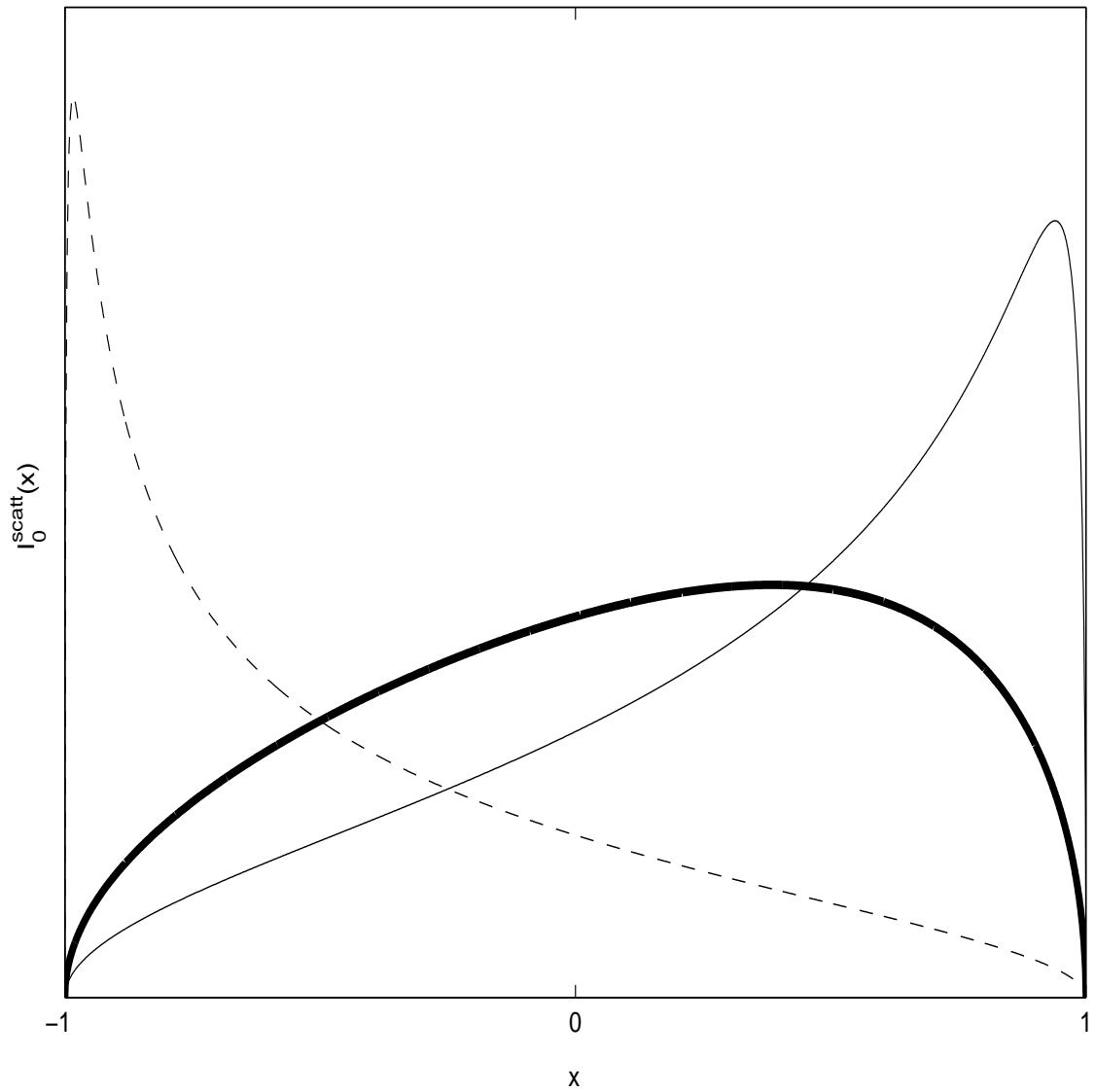


Figure 1: The thick line: $\Delta_0 = 0.2$, the thin line: $\Delta_0 = 0.7$, the dash line: $\Delta_0 = -1.2$.